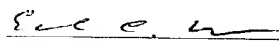


JC20 Rec'd PCT/PTO SEP 20 2001

FORM PTO-1390 (REV. 11-2000)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE	ATTORNEY'S DOCKET NUMBER 31180.830018.000
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371			U.S. APPLICATION NO. (If known, see 37 CFR 1.5) 09/937116
INTERNATIONAL APPLICATION NO. PCT/AU00/00206	INTERNATIONAL FILING DATE March 17, 2000	PRIORITY DATE CLAIMED March 22, 1999	
TITLE OF INVENTION PROCESS AND APPARATUS FOR THE CONVERSION OF CARBONACEOUS MATERIALS			
APPLICANT(S) FOR DO/EO/US Trevor Redvers BRIDLE and Stefan SKRYPISKI-MANTELE			
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:			
<ol style="list-style-type: none"> 1. <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a filing under 35 U.S.C. 371. 2. <input type="checkbox"/> This is a SECOND or SUBSEQUENT submission of items concerning a filing under 36 U.S.C. 371. 3. <input checked="" type="checkbox"/> This is an express request to begin national examination procedures (35 U.S.C. 371(f). The submission must include items (5), (6), (9) and (21) indicated below. 4. <input type="checkbox"/> The US has been elected by the expiration of 19 months from the priority date (Article 31). 5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 37(c)(2)) <ol style="list-style-type: none"> a. <input checked="" type="checkbox"/> is attached hereto (required only if not communicated by the International Bureau). b. <input type="checkbox"/> has been communicated by the International Bureau c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US). 6. <input checked="" type="checkbox"/> An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)). <ol style="list-style-type: none"> a. <input checked="" type="checkbox"/> is attached hereto. b. <input type="checkbox"/> has been previously submitted under 35 U.S.C. 154(d)(4). 7. <input type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)). <ol style="list-style-type: none"> a. <input type="checkbox"/> are attached hereto (required only if not communicated by the International Bureau). b. <input type="checkbox"/> have been communicated by the International Bureau. c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired. d. <input type="checkbox"/> have not been made and will not be made. 8. <input type="checkbox"/> An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371 (c)(3)). 9. <input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)). 10. <input type="checkbox"/> An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)). 			
Items 11 to 20 below concern document(s) or information included:			
<ol style="list-style-type: none"> 11. <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98. 12. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included. 13. <input type="checkbox"/> A FIRST preliminary amendment. 14. <input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment. 15. <input checked="" type="checkbox"/> A substitute specification. 16. <input type="checkbox"/> A change of power of attorney and/or address letter. 17. <input type="checkbox"/> A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825. 18. <input type="checkbox"/> A second copy of the published international application under 36 U.S.C. 19. <input type="checkbox"/> A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4). 20. <input checked="" type="checkbox"/> Other items or information: International Preliminary Examination Report 			

JC03 Rec'd PCT/PTO 20 SEP 2001

I/S/ Application No. (If known, see 37 CFR 1.5) 09/937116		INTERNATIONAL APPLICATION NO. PCT/AU00/00206		ATTORNEY'S DOCKET NUMBER 31180.830018.000	
21. <input checked="" type="checkbox"/> The following fees are submitted:				CALCULATIONS PTO USE ONLY	
BASIC NATIONAL FEE (37 CFR 1.492(A) (1) - (5)):					
Neither international preliminary examination fee (37 CFR 1.492) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO				\$1000.00	
International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search report prepared by the EPO of JPO				\$860.00	
International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO				\$710.00	
International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4)				\$690.00	
International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)(4)				\$100.00	
ENTER APPROPRIATE BASIC FEE AMOUNT =				\$1000.00	
Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input checked="" type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).				\$ 130.00	
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	\$	
Total claims	31- 20 =	11	x \$18.00	\$198.00	
Independent claims	10 - 3 =	7	x \$80.00	\$560.00	
MULTIPLE DEPENDENT CLAIM(S) (if applicable)				+ 270.00	
TOTAL OF ABOVE CALCULATIONS =				\$2158.00	
<input checked="" type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by 1/2.				+ \$1079.00	
SUBTOTAL =				\$1079.00	
Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFT 1.492(f)).				\$	
TOTAL NATIONAL FEE =				\$	
Fee for recording the enclosed assignment (37 CFR 1.2(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property				+ \$	
TOTAL FEES ENCLOSED =				\$1079.00	
				Amount to be refunded: \$	
				charged: \$	
a. <input type="checkbox"/> A check in the amount of \$ _____ to cover the above fees is enclosed. b. <input checked="" type="checkbox"/> Please charge my Deposit Account No. <u>08-2623</u> in the amount of \$ _____ to cover the above fees. c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <u>08-2623</u> . A duplicate copy of this sheet is enclosed. d. <input type="checkbox"/> Fees are to be charged to a credit card. WARNING: Information on this form may become public. Credit card information should not be included on this form. Provide credit card information and authorization on PTO-2038.					
NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.					
SEND ALL CORRESPONDENCE TO:					
Brian P. Kinnear Holland & Hart LLP P.O. Box 8749 Denver, Colorado 80201			 SIGNATURE		
			<u>Earl C. Hancock</u> NAME		
			<u>19,472</u> REGISTRATION NUMBER		

2/18/15

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Atty. Docket No.: 31180.830018.0000

Title

"Process and Apparatus for the Conversion of Carbonaceous Materials"

Field of the Invention

5 The present invention relates to a process and apparatus for the conversion of carbonaceous materials. More particularly, the process and apparatus of the present invention provides an improvement to the oil product of the conversion of the organic components of sewage and industrial sludges.

Discussion of the Prior Art

10 Sewage sludge is an unavoidable by-product of the treatment of sewage and other wastewaters. Traditionally, disposal of such sludge is expensive and typically constitutes half of the total annual costs of wastewater treatment. Historically, the major sludge disposal options have included agricultural utilisation, landfilling and incineration. Also historically, wastewater treatment plants have been designed to minimise sludge production and most effort is expended to stabilise and reduce the sludge volume prior to disposal or utilisation.

20 The solids component of sewage sludge comprises a mixture of organic materials composed of mostly crude proteins, lipids and carbohydrates. These solids further comprise inorganic materials such as silt, grit, clay and lower levels of heavy metals. For example, a typical raw sewage sludge comprises approximately 50 to 90% volatile matter and 25 to 40% organic carbon. Some sewage sludges already exceed current land application contaminant standards and consequently cannot be used agriculturally or are classified hazardous waste, largely due to their organochlorine content.

25 Many sludge processing options have been proposed in the past. Such options have the potential to convert a fraction of the organic material into usable energy and even less have been demonstrated as viable net energy producers

at full scale. One common process involves anaerobic digestion of sewage sludge in which approximately 25% of available organic materials is converted to produce a gas rich in methane. Historically, other alternatives have included starved air incineration and gasification.

- 5 A significant problem associated with the above processes relates to the fact that the principle usable energy-containing products are gases which are generally not easily condensable and are of a low net energy content. Accordingly, such gases are impossible or uneconomic to store and must generally be used immediately. Further, it is generally only practicable to use
10 them to produce relatively low grade energy, such as steam, and flare them to waste during periods of little or not demand. Not surprisingly, it is preferable that any process used result in storable (liquid or solid), transportable and if possible, upgradable energy-containing products. Such products would include synthetic oils. It is consequently desirable that there be optimum
15 production of storable energy having any non-storable products, used in the operation of the process itself.

Disposal of sewage sludge has become more problematic recently due to the fact that:

- (a) agricultural use of sewage sludge is restricted by its contaminant
20 content, particularly the organochlorine content, and within this group the dioxins have become the limiting factor,
- (b) ocean disposal is banned,
- (c) landfilling is to shortly be banned in the European Union; and
- (d) incineration of sewage sludge is opposed by the public primarily with
25 respect to the dioxin issue (reformation of dioxin during hot flue gas cooling). Consequently recent research work on thermal sludge disposal processes concentrates on control of organochlorine compounds across the process.

In US Patents 4618735 and 4781796, there is described a process and apparatus for the conversion of sludges by heating and chemical reaction in order to obtain useful storable products therefrom, including oils. The process comprises the steps of heating dried sludge in a heating zone in the
5 absence of oxygen to a temperature of at least 250°C for the volatilization of oil producing organic material therein, resulting in heating zone gaseous products and sludge residue, removing the said gaseous product from the heating zone; thereafter contacting heated sludge residue in a reaction zone with the removed heating zone gaseous products in the absence of oxygen at a
10 temperature of 280°C to 600°C for repeated intimate gas/solid contact at temperatures sufficient to cause gas/solid contact, oil producing reactions to occur within the heating zone, gaseous products catalysed by the heated sludge residue resulting in reaction zone gaseous products containing oil products; removing the reaction zone gaseous products from the reaction zone
15 and separating at least the condensable oil products therefrom.

Also disclosed is an apparatus for the conversion of sludge, said apparatus comprising an enclosure establishing a heated heating zone having an inlet thereto for dried sewage sludge and separate outlets therefrom for heating zone gaseous products and residual heating zone solid products; conveyor
20 means within the heating zone enclosure for conveying solid products from its inlet to its solid products outlet; and enclosure establishing a heated reaction zone having separate inlets thereto for gaseous and solid products and separate outlets therefrom for gaseous and solid products; conveyor means within the reaction zone enclosure for conveying solid products from its solid
25 products inlet to its solid products outlet; a heating zone solid products outlet being connected to the reaction zone solid products inlet for the passage of solid products between them; and duct means connecting the heating zone gaseous products outlet to the reaction zone gaseous products inlet.

In US Patents 5847248 and 5865956 there is disclosed a process and apparatus based on the process and apparatus of US Patents 4618735 and 4781796, with the following improvements.

5 The gaseous products from the heating zone are transferred to either an indirect or direct condenser with oil/water separation. The resulting oil and/or non-condensable products are injected into a second reactor. Sludge residue or char from the first reactor is transferred to the second reactor by way of a transfer line. The transfer line is equipped with a valve system to ensure that no gaseous products by-pass the condensation system.

10 In the second reactor, provided with heating means, the heated sludge residue from the first reactor is contacted with the revaporised oil or oil and non-condensable gaseous products from the condensation system in the absence of oxygen at a maximum temperature of 550°C. Such allows reductive, heterogenic, catalytic gas/solid phase reactions for the generation of clean
15 products and high quality oil product. A conveyor and motor is provided to move the solid product or char through the second reactor.

Gaseous products are subsequently removed from the second reactor for passage through a further condenser and oil/water separation system or for ducting to a burner for direct combustion. In the case of passage through a
20 further condenser and oil/water separation system a volume of non-condensable gaseous product, a volume of reaction water and a volume of refined, low viscosity oil is produced. Solid products or char are removed from the second reactor by way of a further transfer line having provided therein a screw conveyor for ensuring both no air ingress into and no gaseous
25 product egress from the second reactor. The screw conveyor is connected to a cooling system to cool the solid product or char to less than 100°C before discharge to atmosphere.

The process and apparatus for the conversation of carbonaceous materials of the present invention has as one object thereof to provide a more simple and

In accordance with the present invention there is provided a process for the
5 conversion of sewage sludges, the process characterised by the steps of:

- (a) feeding dried sludge through a reactor;
- (b) heating the dried sludge in the reactor in the absence of oxygen for the volatilization of oil producing organic materials therein, resulting in gaseous products and sludge residue;
- (c) transferring the gaseous products from the reactor to a catalytic converter;
- (d) contacting the gaseous products from the reactor or the reheated oil and/or non-condensable products, if any, with a catalyst in the catalytic converter in the absence of oxygen;
- (e) removing the gaseous products from the catalytic converter; and
- (f) condensing and oil/water separating the gaseous products of the catalytic converter.

Preferably, sludge residue from the reactor is transferred to a storage bin through a valve system for ensuring both no air ingress into and no gaseous product egress from the reactor.

Still preferably, the feeding of the dried sludge through the reactor utilises a
25 feed system that ensures both no air ingress into, and no escape of gaseous
products from, the reactor.

The temperature of the reactor is preferably at least 250°C. The temperature of the reactor is still preferably about 450°C.

- 5 (g) transferring the gaseous products from the reactor to a condensation system to condense the oil product from the gaseous products; and
- (h) reheating water free oil and/or non-condensable products, if any, from the condensation system in an oil reheater.

The condensation system of step (f) preferably comprises a direct condenser. The condensation of step (g) preferably comprises indirect condensation at 10 $\geq 100^{\circ}\text{C}$.

The direct transfer of gaseous products of step (b) from the reactor to the catalytic converter preferably takes place in heat traced lines.

The temperature of the catalytic converter is preferably up to 650°C, thereby promoting conductive, catalytic gas/solid phase reactions and substantially eliminating hetero-atoms, including nitrogen, oxygen, sulphur, and halogens. The catalytic converter temperature is preferably in the range of 400 to 550°C. Still preferably, the catalytic converter temperature is in the range of 400 to 420°C.

The catalytic converter may contain a catalyst, the catalyst being chosen from
20 any of zeolite, activated alumina, γ -aluminum oxide, silicon oxide and oxides
of alkali, earth alkali and transition metals. Preferably, the catalyst is zeolite.

The process of the present invention may be still further characterised by the step of testing the miscibility of the oil product with a hydrocarbon solvent, for example diesel fuel. In response, the catalytic converter conditions may be modified, thereby optimizing the catalytic conversion of sewage sludge, particularly the elimination of hetero-atoms such as halogens, nitrogen, oxygen and sulphur.

5 In accordance with the present invention there is further provided a process for the optimisation of the process for the conversion of sewage sludges as set out above with particular relevance to the removal of hetero-atoms to produce a product miscible with a hydrocarbon solvent such as diesel fuel. Oils miscible in a hydrocarbon solvent have lower viscosity, water content and hetero-atom content, i.e. a higher oil quality. Consequently the miscibility with a hydrocarbon solvent can be used as an analytical tool to optimize the process.

10 The present invention still further provides a process for the conversion of sewage sludges, the process characterised by the steps of:

- (a) feeding dried sludge through a first reactor;
- (b) heating the dried sludge in the first reactor in the absence of oxygen for the volatilization of oil producing organic materials therein, resulting in gaseous products and
15 sludge residue;
- (c) transferring gaseous products from the first reactor to a first condensation system;
- (d) transferring sludge residue to a second reactor where it is heated with oil and/or non-condensable products from the
20 first condensation system;
- (e) transferring the gaseous products of the second reactor to a catalytic converter;
- (f) removing the gaseous products from the catalytic converter; and
- 25 (g) condensing and oil/water separating the gaseous products of the catalytic converter.

Preferably, the temperature of both reactors is about 450°C. Still preferably, the catalytic converter has a temperature of about 400 to 420°C.

30 In accordance with the present invention there is still further provided an apparatus for the conversion of carbonaceous materials, the apparatus

characterised by a feed system for dried material to be conveyed, a reactor, and a catalytic converter, the reactor having a solid product discharge outlet and a transfer line provided for transport of gaseous product directly or indirectly to the catalytic converter.

- 5 Preferably, a first condensation system is provided in-line between the reactor and catalytic converter. The first condensation system preferably includes an oil/water separation system.

The catalytic converter is preferably adapted to contact heated catalyst contained therein with oil or oil and non-condensable products of the
10 condensation system, wherein gaseous products may be removed from the catalytic converter. A reheater may be provided between the first condensation system and the catalytic converter.

A second condensation system is preferably provided to receive gaseous product from the catalytic converter.

15

Description of the Drawings

The present invention will now be described, by way of example only, with reference to two embodiments thereof and the accompanying drawings, in which:

20

Figure 1 is a schematic diagram of an apparatus for the conversion of carbonaceous materials in accordance with a first embodiment of the present invention; and

Figure 2 is a schematic diagram of an apparatus for the conversion of carbonaceous materials in accordance with a second embodiment of the present invention.

Best Mode(s) for Carrying Out the Invention

In Figure 1 there is shown an apparatus 10 for the conversion of sludges in accordance with a first embodiment of the present invention. The apparatus comprises a feed system 12 for dried sludge 14. The feed system 12 feeding
5 said sludge 14 to a reactor 16. The feed system 12 is such that it ensures both no air ingress and no gaseous egress from the reactor 16.

The reactor 16 is provided with heating means 18 thereabout, such that dried sludge may be heated therein in the absence of oxygen to at least 250°C. This results in the volatilization of oil producing organic materials in the reactor
10 16. Further, this reaction provides gaseous products and sludge residue or char. A screw conveyor 20 and a motor 22, or their equivalent, is provided to move the sludge through the reactor 16.

The gaseous products from the reactor 16 are either transferred to a combustion system 24, to a catalytic converter 26 or to a first condensation
15 system 28 with oil/water separation. Further, the first condensation system 28 may comprise either a direct or an indirect condenser. The resulting oil is injected, via line 30, with or without any non-condensable products, via line 32, from the first condensation system 28 into an oil reheater 34 where the oil and/or non-condensable products 32 are heated in the absence of oxygen to a
20 maximum temperature of 650°C.

The gaseous products from the reactor 16 or the preheated oil and/or non-condensable products from the oil reheater 34 are injected into the catalytic converter 26, via lines 31 and 33 respectively. Line 31 from the reactor 16 to the catalytic converter 26 is heat traced. The catalytic converter 26 is
25 provided with a heating means 36 thereabout such that the preheated oil and/or non-condensable products may be heated therein in the absence of oxygen to a maximum temperature of 650°C. This allows reductive, catalytic removal of hetero-atoms to produce an oil product at a viscosity that is lower than that of diesel. The refined gaseous products from the catalytic converter

Run 2: Conversion of raw sludge from the Atlanta WWTP, otherwise as per Run 1.

Run 3: Subiaco sludge processed in accordance with the second embodiment of the present invention with gaseous products being condensed and the oil product, after passing through the reheater prior to the second reactor before being contacted with the catalyst (aluminum oxide impregnated with transition metal oxides) in the catalytic converter. The catalytic converter temperature was maintained at 400°C.

Run 4: As per Run 3 but using Atlanta sludge, with gaseous products being directly transferred to the catalytic converter.

The above results clearly show that the oil viscosity and water content are reduced significantly by the use of the catalytic converter, for example compare Runs 1 and 3, and 2 and 5.

5 A second series of tests was conducted to determine the influence of catalyst support. Test results are shown in Table 2.

Table 2 - Oil Quality versus Catalyst Conversion¹

Run No. Parameter	Run 6	Run 7	Run 8	Run 9	Run 10	Run 11
Viscosity @ 40°C [cSt]	25.8	29.8	3.2	7.7	7.7	7.7
Water Content [%]	9.4	6.7	0.72	2.2	2.2	2.2

Run 6: Atlanta sludge was processed in accordance with the second embodiment of the present invention, with gaseous products being condensed and the oil product being preheated prior to injection into the second reactor from where it is transferred to the catalytic converter. The temperature of both reactors and catalytic converter was controlled at 450°C. Sludge feed rate was

controlled at 600 g/hr. Pure Al_2O_3 was used as a catalyst and was activated at 600°C.

- Run 7: As per Run 6, with Al_2O_3 being activated at 1200°C as a catalyst.
- Run 8: As per Run 6, with Al_2O_3 being impregnated with transition metal oxides and catalytic converter temperature being 400°C.
- Run 9: As per Run 6, using silica gel as catalyst.
- Run 10: As per Run 6, processing Subiaco sludge and using zeolite as catalyst with catalytic converter temperature being 420°C.
- Run 11: As per Run 6, using char as catalyst.

The results show that pure Al_2O_3 is not as good as Al_2O_3 doped with transition metals. Silica gel and zeolite appear to be good catalysts and the conversion char is not as good as Al_2O_3 as a catalyst.

- 5 A third series of tests were conducted to assess the impact of catalyst temperature on oil quality and yield. Results are shown in Table 3.

Table 3 - Oil Quality and Yield versus Catalyst Temperature

Run No. Parameter	Run 12	Run 13	Run 14	Run 15	Run 16	Run 17	Run 18	Run 19	Run 20
Viscosity @ 40°C [cSt]	12.4	8.9	31.7	28.4	26.2	30.3	26.8	16.5	16.4
Water Content [%]	23.3	17.9	25.1	25.3	22.8	22.5	22.0	21.4	19.6

- Run 12: Subiaco sludge was processed in accordance with the first embodiment of the present invention, with gaseous products being condensed and the oil product being preheated prior to injection

- 5 A fourth series of tests were conducted to assess the impact of catalyst to oil vapor ratio on oil quality. Results are shown in Table 4.

Table 4 – Catalyst to oil vapour ratio versus oil Quality and Quantity

Run No. Parameter	Run 21	Run 22	Run 23
Weight Hourly Space Velocity (WHSV) [hr ¹]	2.3	1.1	0.7
Viscosity @ 40°C [cSt]	13	4.5	4.8
Water Content [%]	3.6	1.2	1.1
Oil Yield [%]	23.2	18.8	18.3

Run 21: Atlanta sludge was processed in accordance with the second embodiment of the present invention, with gaseous products being condensed and the oil product being preheated prior to injection into the second reactor from where it is transferred to the catalytic converter. The temperature of both reactors and the catalytic converter were controlled at 450°C. Sludge feed rate was controlled at 600 g/hr. Aluminum oxide impregnated with transition metal oxides was used as a catalyst and 80g of catalyst used.

Run 22: As per Run 21, with 165g of catalyst being used.

Run 23: As per Run 21, with 265g of catalyst being used.

- 5 As expected, the results show improved oil quality at lower WHSV (higher catalyst mass), albeit at a reduced oil yield. There does not appear to be any further improvement in oil quality by reducing the WHSV to below 1 hr.¹

A fifth series of tests was conducted to assess the impact of catalyst regeneration on oil quality. Results are shown in Table 5.

Table 5 – Catalyst Regeneration versus Oil Quality

Run No. Parameter	Run 24	Run 25	Run 26	Run 27	Run 28	Run 29
Viscosity @ 40°C [cSt]	11.8	17.0	14.6	17.5	20.5	20.2
Oil Yield [%]	16.8	20.6	19.7	16.8	17.1	19.2

Run 24: Subiaco sludge was processed in accordance with the first embodiment of the present invention, with gaseous products being transferred directly to the catalytic converter. The temperature of the reactor and the catalytic converter was controlled at 450 and 550°C respectively. Sludge feed rate was controlled at 600g/hr. 210g of native zeolite was used as a catalyst.

Run 25: As per Run 24, with the catalyst being regenerated on-line for the first time.

Run 26: As per Run 24, with the catalyst being regenerated on-line for the second time.

Run 27: As per Run 24, with the catalyst being regenerated on-line for the third time.

Run 28: As per Run 24, with the catalyst being regenerated on-line for the fourth time.

Run 29: As per Run 24, with the catalyst being regenerated on-line for the fifth time.

The results show a slight deterioration in catalyst efficacy via on-line
5 regeneration. However, performance appeared to stabilise after 3 or 4
regenerations.

A sixth series of tests were conducted to compare oil yield and quality from the dual reactor system with intermediate condensation and the single reactor system without intermediate condensation. The results are shown in Table 6.

Table 6 – Oil Quality Data: Impact of Dual Versus Single Reactor Systems

5

Parameter \ Run No.	Run 30	Run 31
Oil Yield [%]	18.8	16.9
Viscosity [cSt]	4.5	7.6

Run 30: Atlanta WWTP sludge was processed in accordance with the second embodiment of the present invention, with gaseous products being condensed after the first reactor and the oil product was injected into the second reactor. The temperature of both the reactor and the catalytic converter was controlled at 450°C. Sludge feed rate was controlled at 600g/hr. Al_2O_3 was used as the catalyst.

Run 31: As per Run 30 but rather in accordance with the first embodiment of the present invention, that is without the intermediate condensing of gaseous products or the second reactor.

10 These results indicate that the presence of the second reactor, and the condensation of gaseous products before injection thereto, does not have a significant impact upon oil yield and quality, as might otherwise have been expected, when compared with the apparatus 10 utilising the single reactor 16.

A seventh series of tests were conducted to determine the impact of intermediate condensation on oil quality and yield in the single reactor system. Tests results are shown in Table 7.

Table 7 – Oil Data: Impact of Intermediate Condensing

Run No. Parameter	Run 13/32	Run 14
Oil Viscosity [cSt]	8.9	31.7
Oil Yield [%]	17.9/23.3	25.1

5

Runs Subiaco sludge was processed in accordance with the first
13/32: embodiment of the present invention, with gaseous products being condensed and the oil reheated before being injected to the catalytic converter. The temperature of the reactor was maintained at 450°C and the converter at 420°C. The catalyst was zeolite.

Run 14: As per Run 13/32 with gaseous products being transferred directly to the catalytic converter which as operated at 450°C.

These results indicated that the intermediate condenser has a significant impact on oil quality. There is however, a significant oil yield penalty when using the intermediate condenser.

10 An eighth series of tests was conducted to determine the temperature at which the catalytic converter must be operated to achieve acceptable results in a single reactor system, without intermediate condensation. Test results are shown in Table 8.

Table 8– Oil Quality Data: Impact of Catalytic Converter Temperature

Run No.	Run 32	Run 33
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Parameter		
Weight of Catalyst [g]	180	210
Catalyst Temperature [°C]	420	550
Oil Yield [%]	23.3	15.7
Viscosity [cSt]	8.9	11.8

Run 32: Subiaco sludge was processed in accordance with the first embodiment of the present invention, with gaseous products being condensed after the single reactor prior to injection to the catalytic converter. The temperature of the reactor and the catalytic converter was controlled at 450°C and 420°C. Sludge feed rate was controlled at 600 g/hr. Zeolite was used as the catalyst.

Run 33: As per Run 32 but without the condensing of gaseous products prior to introduction to the catalytic converter, which was operated at 550°C.

These results indicate that the condensing of the gaseous products of the single reactor prior to injection to the catalytic converter provides an improved oil quality and yield when compared with not having conducted the condensation step, when using zeolite as the catalyst.

These results further indicate that the catalytic converter must be operated at about 130°C higher if it is desired to produce oil of the same quality without intermediate condensation of the gaseous products of the reactor. As seen in Table 8, this produces a significant drop in oil yield.

10 The process for the conversion of sewage sludges of the present invention may be optimised with regard to removal of hetero-atoms through use of miscibility with a standard hydrocarbon source such as diesel fuel. It is apparent that the miscibility with diesel fuel is significantly influenced by the presence of hetero-atoms such as oxygen, nitrogen and sulphur. Such hetero-

atoms particularly oxygen and sulphur and to a lesser degree nitrogen are removed by the catalytic converter generating an oil that is miscible at any ratio with diesel fuel. Accordingly, the miscibility of the oil product is a direct measurement of the quality of the oil. The miscibility can be directly
5 correlated with viscosity, meaning the better mixing the lower the viscosity. As stated in US Patent 5847248, viscosity can also be used as an indicator for the destruction rate of organochlorine compounds and consequently, since the heavy metal oxides present in the catalytic converter area the same as in sewage sludge, the dehalogenation of organochlorines will take place in the
10 catalytic converter at a much higher rate than in the second reactor of US Patents 5847248 and 5865956, and miscibility can also be used as a control tool for organochlorine destruction.

The process and apparatus of the present invention provide reductive, thermal heterogenic catalytic solid/gas phase reactions in order to obtain storable
15 products with unrestricted use.

Modifications and variations such as would be apparent to the skilled addressee are considered to fall within the scope of the present invention.

CLAIMS

1. A process for the conversion of sewage sludges, the process characterised by the steps of:
 - (a) feeding dried sludge through a reactor;
 - 5 (b) heating the dried sludge in the reactor in the absence of oxygen for the volatilization of oil producing organic materials therein, resulting in gaseous products and sludge residue;
 - (c) transferring the gaseous products from the reactor to a catalytic converter;
 - 10 (d) contacting the gaseous products from the reactor or the reheated oil and/or non-condensable products, if any, with a catalyst in the catalytic converter in the absence of oxygen;
 - (e) removing the gaseous products from the catalytic converter; and
 - (f) condensing and oil/water separating the gaseous products of the
15 catalytic converter.
2. A process according to claim 1, wherein sludge residue from the reactor is transferred to a storage bin through a valve system for ensuring both no air ingress into and no gaseous product egress from the reactor.
- 20 3. A process according to claim 1 or 2, wherein the feeding of the dried sludge through the reactor utilises a feed system that ensures both no air ingress into, and no escape of gaseous products from, the reactor.
4. A process according to claim 2 or 3, wherein the temperature of the reactor is at least 250°C.
- 25 5. A process according to claim 4, wherein the temperature of the reactor is about 450°C.
6. A process according to any one of the preceding claims, the process further characterised by the method steps of:

- (g) transferring the gaseous products from the reactor to a condensation system to condense the oil product from the gaseous products; and
- (h) reheating water free oil and/or non-condensable products, if any, from the condensation system in an oil reheater.
- 5
7. A process according to any one of the preceding claims, wherein the condensation system of step (f) comprises a direct condenser.
8. A process according to claim 6, wherein the condensation of step (g) comprises indirect condensation at $>100^{\circ}\text{C}$.
- 10 9. A process according to any one of the preceding claims, wherein the direct transfer of gaseous products of step (b) from the reactor to the catalytic converter takes place in heat traced lines.
10. A process according to any one of the preceding claims, wherein the temperature of the catalytic converter is up to 650°C , thereby
- 15 promoting reductive, catalytic gas/solid phase reactions and substantially eliminating hetero-atoms, including nitrogen, oxygen, sulphur, and halogens.
11. A process according to claim 10, wherein the catalytic converter temperature is in the range of 400 to 550°C .
- 20 12. A process according to claim 10, wherein the catalytic converter temperature is in the range of 400 to 420°C .
13. A process according to any one of the preceding claims, wherein the catalytic converter contains a catalyst, the catalyst being chosen from any of zeolite, activated alumina, γ -aluminium oxide, silicon oxide and
- 25 oxides of alkali, earth alkali and transition metals.
14. A process according to claim 13, wherein the catalyst is zeolite.

15. A process according to any one of claims 6 to 14, wherein the process further comprises the step of testing the miscibility of the oil product with a hydrocarbon solvent and modifying the conditions of the catalytic converter in response thereto.
- 5 16. A process according to any one of claims 6 to 15, wherein the oil product of the process is miscible with a hydrocarbon solvent.
17. A process according to claim 16, wherein the solvent is diesel fuel.
18. A process for the conversion of sewage sludges, the process characterised by the steps of:
- 10 (a) feeding dried sludge through a first reactor;
- (b) heating the dried sludge in the first reactor in the absence of oxygen for the volatilization of oil producing organic materials therein, resulting in gaseous products and sludge residue;
- (c) transferring gaseous products from the first reactor to a first condensation system;
- 15 (d) transferring sludge residue to a second reactor where it is heated with oil and/or non-condensable products from the first condensation system;
- (e) transferring the gaseous products of the second reactor to a catalytic converter;
- 20 (f) contacting the gaseous products of step (e) with a catalyst in the catalyst converter in the absence of oxygen;
- (g) removing the gaseous products from the catalytic converter; and
- (h) condensing and oil/water separating the gaseous products of the catalytic converter.
- 25 19. A process according to claim 18, wherein the temperature of both reactors is about 450°C.

ABSTRACT

A process for the conversion of sewage sludges, the process characterized by the steps of: (a) feeding dried sludge through a reactor (16); (b) heating the dried sludge in the reactor (16) in the absence of oxygen for the volatilization of oil producing organic materials therein, resulting in a gaseous products and sludge residue; (c) transferring the gaseous products from the reactor (16) to catalytic converter (26); (d) contacting the gaseous products from the reactor (16) or the reheated oil and/or non-condensable products, I any, with a catalyst in the catalytic converter (26) in the absence of oxygen; (e) removing the gaseous products from the catalytic converter (26); and (f) condensing and oil/water separating the gaseous products of the catalytic converter (26).

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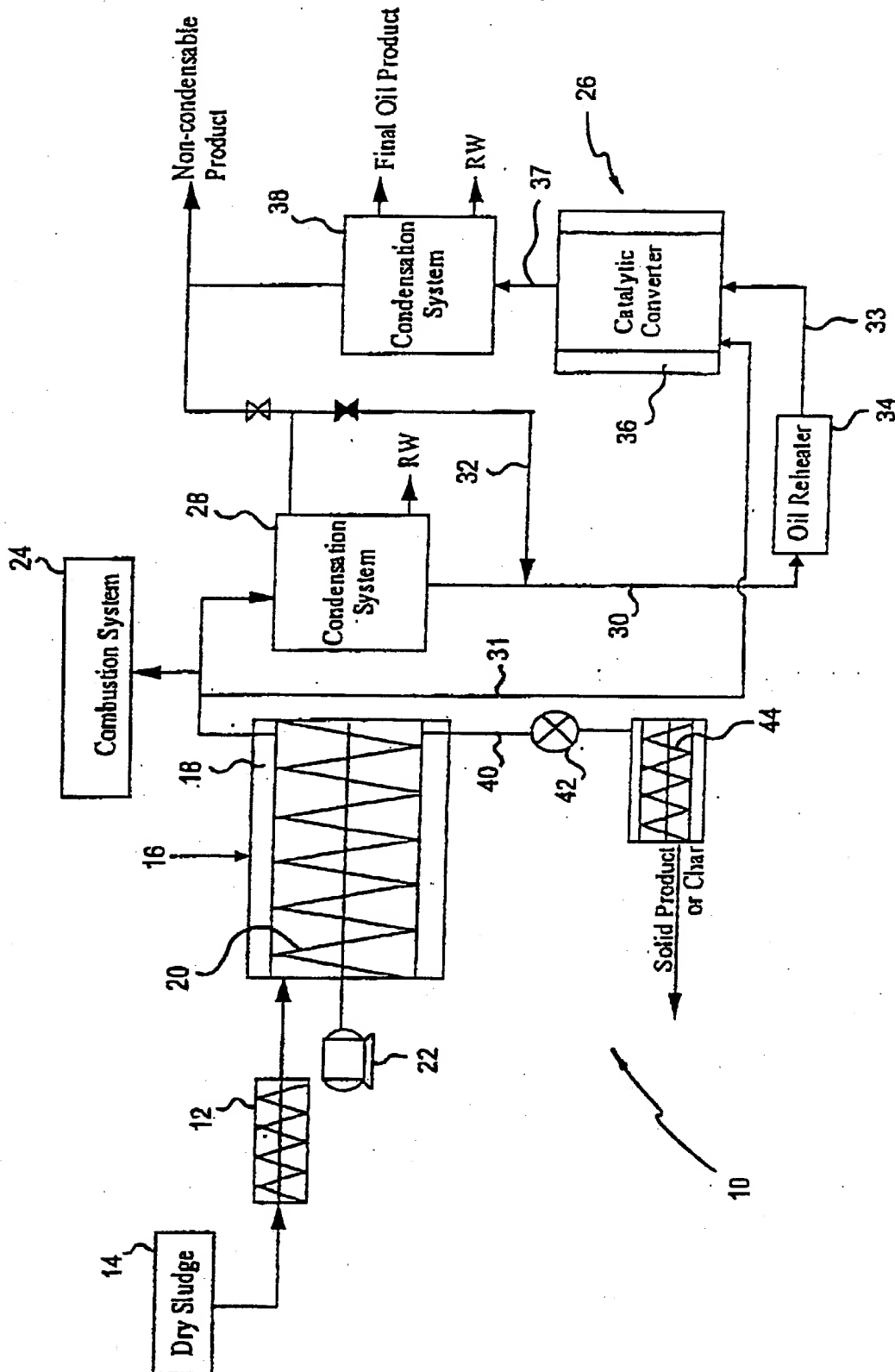


Fig. 1

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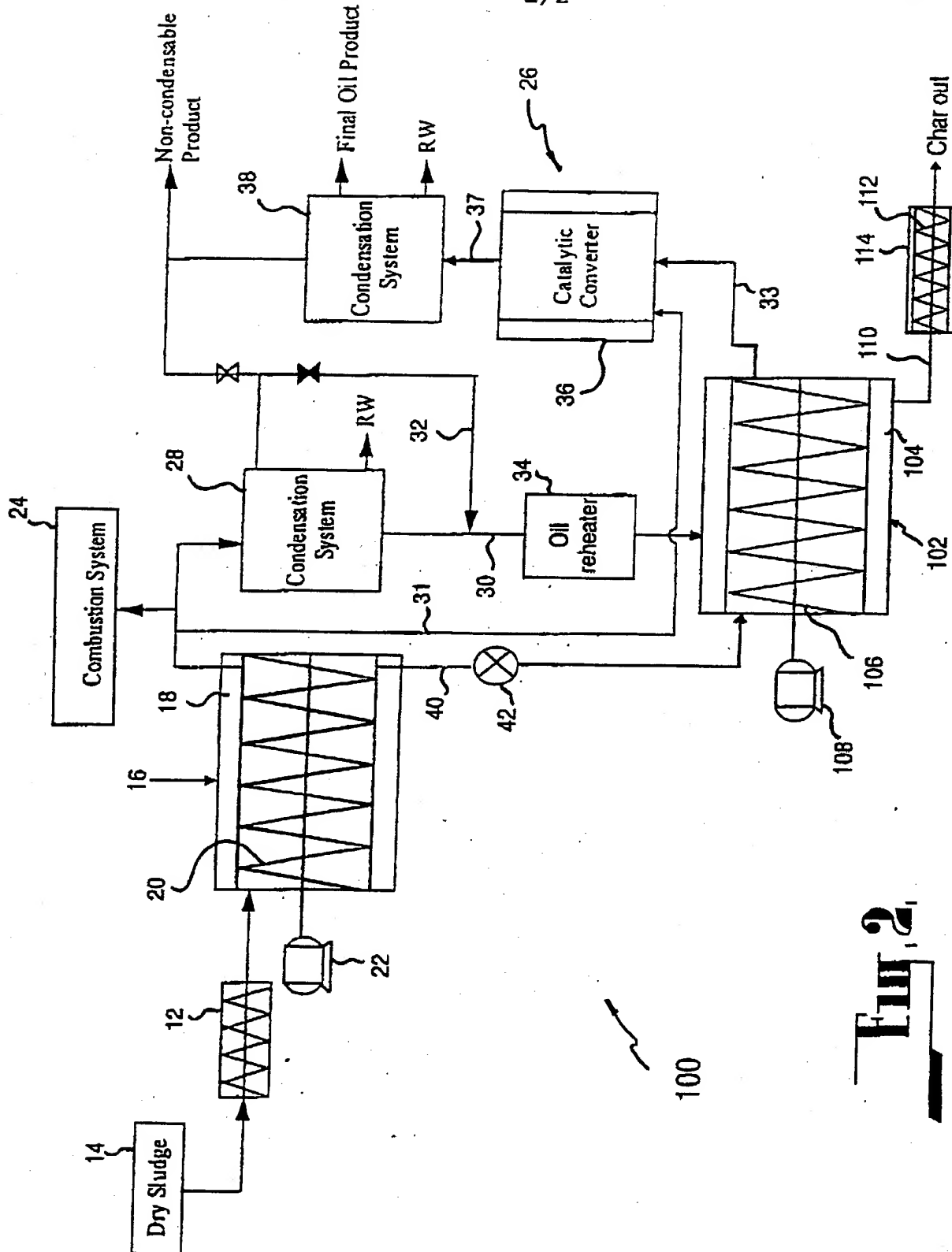


Fig. 2

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PATENT

Attorney Docket No. 31180.830018.000

Express Mail No. EL353030033US

**COMBINED DECLARATION AND POWER
OF ATTORNEY FOR PATENT APPLICATION**

Wray

DECLARATION:

As the below named inventors, we hereby declare that:

Our residences, post office addresses and citizenships are as stated below next to our names.

We believe we are the original, first and joint inventors of the subject matter that is claimed and for which a patent is sought on the invention entitled **PROCESS AND APPARATUS FOR THE CONVERSION OF CARBONACEOUS MATERIALS**, U.S. Patent Application No. 09/937,116, filed September 20, 2001.

The persons named as the inventors in this application are: **TREVOR REDVERS BRIDLE and STEFAN SKRYPISKI-MANTELE**

We hereby state that we have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

We acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations, § 1.56(a), as attached.

We hereby claim foreign priority benefits under Title 35, United States Code, § 119/365 of any foreign application(s) for patent of inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on the basis of which priority is claimed:

- ☐ no such applications have been filed.
☒ such applications have been filed as follows:

FOREIGN APPLICATION(S), IF ANY, CLAIMING PRIORITY UNDER 35 USC § 119/365			
COUNTRY	APPLICATION NUMBER	DATE OF FILING (day, month, year)	DATE OF ISSUE (day, month, year)
PCT	PCT/AU00/00206	March 17, 2001	Pending
ALL FOREIGN APPLICATION(S), IF ANY, FILED BEFORE THE PRIORITY APPLICATION(S)			
COUNTRY	APPLICATION NUMBER	DATE OF FILING (day, month, year)	DATE OF ISSUE (day, month, year)
Australia	PP9360	March 22, 1999	Pending

We hereby claim the benefit under Title 35, United States Code, § 120/365 of any United States (s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, § 112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, § 1.56(a) which occurred between the filing date of the prior application and the national or PCT international filing date of this application.

U.S. or PCT APPLICATION NUMBER	DATE OF FILING (day, month, year)	STATUS (patented, pending, abandoned)

POWER OF ATTORNEY:

As named inventors, we hereby appoint the following patent attorneys to prosecute this application and transact all business in the Patent Office connected therewith:

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We hereby declare that all statements made herein of our own knowledge are true and that all statements made on information and belief are believed to be true, and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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§ 1.56 duty to disclose information material to patentability.

(a) A patent by its very nature is affected with a public interest. The public interest is best served, and the most effective patent examination occurs when, at the time an application is being examined, the Office is aware of and evaluates the teachings of all information material to patentability. Each individual associated with the filing and prosecution of a patent application has a duty of candor and good faith in dealing with the Office, which includes a duty to disclose to the Office all information known to that individual to be material to patentability as defined in this section. The duty to disclose information exists with respect to each pending claim until the claim is cancelled or withdrawn from consideration, or the application becomes abandoned. Information material to the patentability of a claim that is cancelled or withdrawn from consideration need not be submitted if the information is not material to the patentability of any claim remaining under consideration in the application. There is no duty to submit information which is not material to the patentability of any existing claim. The duty to disclose all information known to be material to patentability is deemed to be satisfied if all information known to

